Purification of used sunflower frying oil with adsorbent mixtures using an active filtration method

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In this study, purification of used frying sunflower oil was investigated using different adsorbents and their mixtures. The effective adsorbents in reducing the free fatty acid (FFA) content, total polar material (TPM) and the p-Anisidine value (AV) as well as in lightening of the colour, when used in single-use, were aluminium oxide (AO), silica gel (SG), activated charcoal (AC) and bleaching earth (BE). Increasing the adsorbent ratio from 3% to 6% improved the quality of the used oil. Among the binary adsorbent mixtures, 6%AO+6%AC reduced FFA content by 79.1% and the AV by 56.8%. The ternary adsorbent mixtures reduced the conjugated diene (CD) content of oil in the following order 3%SG+3%M+3%B, 3%AO+3%AC+6%Z, 3%AO+3%M+3%B and 3%AO+3%M+3%Z, respectively, providing a percentage improvement (PI) in the range of 60-100%. The binary adsorbent mixture reduced the viscosity of the used oil. The highest value was 3%AO+6%SG (42.6%), whereas the following ternary mixtures of 3%AO+3%SG+3%AC (48.4%) and 3%AO+3%SG+3%B (47.6%) were the most successful. The utilisation of adsorbents singly or in either binary or ternary mixtures resulted in increasing the L* value of the used oil and decreasing the a* and b* values. The adsorbents and their mixtures are believed to be useful in the purification of used oil.

Keywords: Active filtration; bleaching earth; used frying oil; adsorbent mixtures; free fatty acid.

Abbreviations

AC (activated charcoal). *AO* (aluminum oxide). *AV* (p-Anisidine value). *B* (bentonite). *BE* (bleaching earth). *BR* (britesorb). *CS* (silverskin). *CSA* (silverskin ash). *FFA* (Free fatty acid content). *FP* (fry powder). *HB* (hubersorb). *M* (magnesol XL). *NCS* (silverskin nanoparticles). *PI* (percentage improvement). *SG* (silica gel). *TCD* (total color difference). *TPM* (total polar material content). *UFPO* (used frying palm oil). *Z* (zeolite).

INTRODUCTION

Deep frying of food products is used extensively both in industrial and in domestic processes owing to several reasons including the specific smell and aroma that the deep-fried product attains and the shortened cooking times. Deep frying provides sensory characteristics of foods (i.e., crispy texture, brownish colour, juicy flavour), however, the negative impacts of frying on foods have been also reported [1-5]. Reactions that take place in oil while frying such as polymerisation, hydrolysis and oxidation occur polar compounds and polymers, which adversely affect human health. The amount of toxic compounds that are formed increases with increasing repeated frying, duration of frying and increasing temperature and the oil finally becomes unusable [6-8].

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Used frying oil has ecotoxic properties polluting the surroundings that it occupies, thus harming organisms that are resident in that environment. Used oil and fat in untreated wastewaters damage other organisms in the environment including the fish when they are discharged into the seas, lakes and rivers through pollution and depletion of oxygen in the water. On the other hand, frying oil contains significant amounts of triglycerides. The interest in the purification of frying oil is a result of several demands regarding environmental awareness, economic reasons and to produce fried foodstuffs with the desired characteristics and to use purified oil in biodiesel production. Many researchers attempted to produce reusable oil through the removal of degradation products in order to purify frying oil. Several methodologies such as active and passive filtration as well as membrane filtration were developed to serve this purpose [7, 9].

Particles of a predetermined size may be removed through passive filtration although the chemical characteristics of the frying oil remain mostly unchanged. Not only particles are removed by active filtration but also the chemical composition of the frying oil changes through the removal of polar and polymer compounds that lower the stability of the frying oil. Active filtration is a method of adsorption, which is commonly used in improving the quality of the used frying oil [9-11].

Several studies are available in literature on the regeneration of used frying oil *via* active filtration. These studies investigated the changes in physical and chemical parameters of used oil when several adsorbents and the mixtures of the combination of these adsorbents are used. Ismail *et al.* [1] evaluated the efficiency of coffee silverskin (CS), CS ash (CSA) and CS nanoparticles (NCS) in the regeneration of used frying palm oil (UFPO). Adsorbents were mixed individually with UFPO at level 4% (w/v) for 60 min. The treatment of UFPO caused reductions in the content of free fatty acids (FFA) ranging from 51.2% to 65.0%. The lowest level of peroxide (2.1 meq/kg) was recorded for UFPO treated NCS. The highest reductions (70.0%) in *p*-anisidine value were observed in UFPO treated with CSA and NCS, respectively. Treatment of UFPO with CS, CSA and NCS lowered the polar content from 13.9% to 6.3%, 4.8% and 3.9%, respectively. Filtration of UFPO with CS, CSA and NCS lowered also the viscosity and colour values of treated UFPO. Mancini-Filho *et al.* [12] reported that bleaching clay, charcoal, magnesium oxide and celite as well as their mixtures effectively improved the dielectric constant, free fatty acid (FFA) content, total polar material (TPM) content and the colour of deep-frying oil. McNeill *et al.* [8] found that silica gel (SG) and active carbon reduced the acid number, peroxide number, TPM content as well as the saturated and the unsaturated carbonyl content

of used oil in addition to lightening its colour. Yates and Caldwell [10] claimed that the adsorbents such as diatomaceous earth, acid activated bleaching earth (BE), active alumina, SG, carbon and synthetic magnesium silicate had different adsorption capacities towards the polar materials in used oil. Maskan and Bağcı [7] reported that a mixture composed of 2% pekmez earth, 3% bentonite (B) and 3% magnesium silicate was the most effective mixture in treating used oil. In another study, Bhattacharya *et al.* [13] reported that active charcoal (AC) dust, aluminium oxide (AO), aluminium hydroxide, active clay, celite, SG, active carbon, frypowder (FP, composed of porous rhyolite and citric acid), britesorb (BR, silicone dioxide) and magnesol (M, magnesium silicate) would be used alone or in binary or quadruple combinations in the regeneration of used oil.

Lin *et al.* [14] reported that a commercial filter additive hubersorb (HB, calcium silicate powder) lowered the FFA content of oil by 84.5% and that FP increased the stability of oil by 38.3% and that M lightened the colour by 46.3%. Using HB and M in a mixture reduced the FFA content by 90.8-93.7% and the TPM by 6.0-17.8% whereas it increased stability by 23.4-24.7%. Also, Miyagi *et al.* [15] proposed that the use of SG together with fewer amounts of magnesium oxide in adsorption processes yielded the most desirable outcome and that a combination of adsorption and membrane processes would be used in the treatment of used oil.

In this study, the effectiveness of using several adsorbents (SG, AO, AC, BE, M, B, and zeolite (Z)) alone and in binary or ternary mixtures in the purification of used frying sunflower oil *via* the method of active filtration was investigated. For this purpose, several physical and chemical properties (FFA content, conjugated diene (CD) content; *p*-Anisidine value (AV), change in the TPM; colour and viscosity) were determined prior to and post adsorption in order to compute improvement ratios based on used oil and fresh sunflower oil.

2. MATERIALS AND METHODS

2.1. MATERIALS

Fresh sunflower oil and used frying oil used in this study was obtained from Beypi Inc. (Bolu, Turkey). Oils were stored in a freezer at -18°C until analysis. The SG 60 (particle size 0.063-0.200 μm), AO 90 (standardised), AC were obtained from Merck (Darmstadt, Germany). Z (particle size < 45 μm) was obtained from Sigma-Aldrich (St. Louis, MO, USA) and B was acquired from AppliChem (Darmstadt, Germany). M was purchased from The Dallas group of America Inc. (Jeffersonville, IN, USA) and it was composed of magnesium silicate (50.86%), cellulose

(41.40%) and FDA approved binder (7.74%). Marsan Adana Margarine and oil facilities kindly provided the BE. Other chemicals that were necessary for the analyses were purchased from Merck (Darmstadt, Germany) and Sigma-Aldrich (St. Louis, MO, USA).

2.2 TREATMENT WITH VARIOUS ADSORBENTS OR ADSORBENT MIXTURES IN A SHAKING BATH

Filtered used frying oil (100 g) was mixed with various adsorbents (SG, AC, Z, B, AO, M and BE) at a ratio of 3% or 6% by weight in a sealed Erlenmeyer flask (250-mL) and kept in shaking water baths with a rotational speed of 150 rpm at 60°C for 30 min. The oil was filtered through Whatman 41 filter paper under vacuum. The filtered oil was transferred into coloured glass bottles. The samples were kept at -18°C until analysis after flushing nitrogen. The changes in the TPM content, FFA content, CD content, AV, the Hunter colour values and the viscosity of the used frying oil were determined before and after treatment. Binary and ternary mixtures of the indicated adsorbents were prepared (1:1, 1:2, 1:1:1 or 1:1:2 by weight) and used in adsorption. These adsorbent mixtures were added into used oil and adsorption was allowed to proceed under the previously stated conditions in the shaking water bath. The changes in the TPM content, FFA content, CD content, AV, the Hunter colour values and the viscosity were also determined before and after treatment. The percentage improvement ratios were also calculated in order to determine the effectiveness of the adsorbent mixtures.

2.3 ANALYTICAL METHODS

2.3.1 Determination of TPM

Polar mono- and diglycerides and FFA are formed as a result of the thermal oxidation and the hydrolysis of triglycerides during the frying process. The number of polar materials was determined using Miroil-Optifry Oil Tester (Miroil Division of Oil Process Systems, Inc., Allentown, PA, USA) at room temperature (25°C). Taking measurements using the polar material determination equipment was preferred owing to the ease of use, reliability and rapidity of analysis. The dielectric constant was measured in the equipment to determine the change in the total polar material content of the oil. TPM content of fresh oil was zero and fresh sunflower oil was used to calibrate the equipment. The polar material contents of fresh, used and adsorbent treated oil were also determined using the chromatographic method (Official method Cd 20-91) proposed by AOCS [17] in order to test the reliability of the measurements. Taking TPM content of fresh oil into consideration, both methods yielded similar results.

2.3.2 Determination of the FFA

FFA content was determined by a modified titrimetric method (Official method Ca 5a-40) stated in AOCS [17]. One g of oil was dissolved in 10 mL of hot 95% ethyl alcohol and following the addition of phenolphthalein indicator, it was titrated with 0.1 N standardized NaOH. The FFA was expressed as % oleic acid.

2.3.3 Determination of the conjugated diene (CD)

CD content of the frying oil prior to and following the treatment with adsorbent was determined in a UV-visible spectrophotometer (Shimadzu UV 1700, Kyoto, Japan) at 232 nm (Official method Ti 1a-64) as stated in AOCS [17]. Approximately 10 mL of iso-octane was added to 0.01 g of oil and the absorbance of the solution was measured in a quartz cuvette at 232 nm using iso-octane as a blank solution. Necessary dilutions were carried out if the absorbance values were out of the range of 0.2-0.8. The CD content was determined by taking the absorbance value, concentration and cuvette thickness into consideration using the following formula:

$$CD\ (\%) = 0.84\ (\ (A/bC) - K_o\ )$$

Where:
A is sample absorbance;
b is cuvette thickness (cm);
C is sample concentration (g/l);
K_o is absorptivity by acid or ester groups (0.07 for esters, 0.03 for acids)

2.3.4 Determination of the AV

AV was determined via recording the absorbance values in a Shimadzu UV-1700 spectrophotometer (Kyoto, Japan) as stated in AOCS [17] method Cd 18-90. Approximately 0.1-0.3 g of oil was dissolved in 10 mL of iso-octane. The absorption (Ab) of this solution was measured at 350 nm using iso-octane as the blank solution. Five mL of this solution was transferred to a bottle. One mL of *p*-Anisidine solution (2.5 g/L, prepared in glacial acetic acid) was added into the bottle. Five mL of iso-octane and 1 mL of *p*-Anisidine solution was used as blank in the spectrophotometer. Both the sample and the blank were left to remain for 10 min and the absorbance (As) of the samples was measured in the spectrophotometer at 350 nm against the blank solution. The calculations were conducted as follows:

$$AV = (10 \times (1.2 \times As\text{-}Ab)\ ) / m$$

Where:
As is absorbance of the oil following reaction with *p*-Anisidine;
Ab is absorbance of oil solution;
m is oil content

2.3.5 Determination of viscosity

The dynamic viscosity of the frying oil prior to and following adsorption was measured at 30°C in mPa.s using And SV-10 Vibro viscometer (AND Co., Japan).

2.3.6 Determination of colour using Hunter colour equipment

a* indicates red-green colour, b* indicates yellow-blue color, and L* indicates the lightness and darkness in the Hunter Lab (Colorflex, Hunter Associates Laboratory, Virginia, USA) colour determination equipment. The L*, a* and b* colour values of the fresh oil, used oil and the oil treated with adsorbents in a shaking water bath were determined. The total colour difference (TCD), which is a combination of parameters L*, a* and b* values, is a colorimetric parameter extensively used to characterise the variation of colour in foods during processing [18]. TCD was calculated as follows:

$$TCD = \sqrt{(L_0{}^*-L^*)^2 + (a_0{}^*-a^*)^2 + (b_0{}^*-b^*)^2}$$

Where:
$L_0{}^*$ (46.9);
$a_0{}^*$ (14.6);
$b_0{}^*$ (60.5) refer to reference values;
i.e. colour parameters of used frying oil;
L^*, a^* and b^* values refer to colour values of adsorbent treated oil.

2.3.7 Determination of the fatty acid composition in fresh and used sunflower oil

Oil samples were methylated according to Lin *et al.* [5]. Briefly, 3 mL of 6% HCl in methanol was added to 50 mg of oil and waited in an oven at 75°C for 2 h. After cooling, 2 mL of hexane was added before centrifugation. The upper phase containing fatty acid methyl esters (FAME) was injected to an Agilent 7890A gas-liquid chromatograph with a fused capillary column (HP-88, 100 m × 0.25 mm i.d., 0.20 μm film thickness; J&W Scientific, Folsom, CA), an autoinjector (Agilent 7683B), a flame ionisation detector and analysed according to Karabulut *et al.* [16]. The injector and detector temperatures were 250°C. The column temperature was held at 140°C for 5 min and ramped to 240°C for 10 min at the rate of 4°C /min. Helium was used as a carrier gas and its total flow rate was 30 mL/min. The split ratio was 1:30. FAME were identified with those of standard mixtures (37 FAME mixture, Sigma-Aldrich Inc., St. Louis, MO), and the results were expressed as the percentage of total peak area.

2.4 EVALUATION OF THE EFFICIENCY OF THE ADSORPTION PROCESS

The efficiency of the adsorption process was given in terms of percentage improvement (PI), which is an index for comparing the quality of the treated oil with that of fresh oil according to Miyagi and Nakajima [9]. PI for each parameter was calculated using the following equation:

$$PI = 100 \, (C_u - C_t) / (C_u - C_f)$$

Where:
C_u, C_f and C_t are the contents or property of each parameter in the used, fresh and adsorbent treated oil.

2.5 STATISTICAL ANALYSIS

The SPSS 11.5 software package was used in the statistical evaluation of the data. The differences in the averages were determined using a one-way analysis of variance (ANOVA) and the results were evaluated in terms of a significance level of 0.05. The differences between the runs were determined by the Duncan test. The analyses were carried out in duplicates.

3. RESULTS AND DISCUSSION

3.1 CHARACTERISTICS OF THE FRESH AND USED FRYING SUNFLOWER OIL

Several physical and chemical properties of used and fresh oil that were tested in the present study (Tab. I). The fatty acid composition of fresh oil included C14:0 (0.08%), C16:0 (6.32%), C16:1 (0.12%), C18:0 (3.68%), C18:1-*trans* (0.03%), C18:1-*cis* (30.0%), C18:2-*trans* (0.27%), C18:2n6-*cis* (58.03%), C18:3n6 (0.26%), C18:3n3 (0.06%), C20:1 (0.15%), C21:0 (0.04%), C22:0+C20:3 (0.65%), and C24:0 (0.32%). The used frying oil was determined as having FFA content of 0.85%, change in TPM as 10.5%, CD content of 0.54% and an AV of 34.4 mmol/kg, which were all higher than their equivalents in fresh oil. The thermal oxidation taking place during frying

Table I - Physical and chemical properties of fresh and used sunflower oil

Property	Fresh oil	Used oil
FFA (%)	0.18±0.00	0.85±0.04
TPM (%)	0.0±0.0	10.5±0.2
CD (%)	0.29±0.00	0.54±0.04
AV (mmol/kg of oil)	10.3±0.0	34.4±1.4
Viscosity (mPa.s) at 30°C	54.6±0.0	64.2±0.1
Color L*	55.2±0.0	46.9±0.2
a*	-3.1 ±0.0	14.6±0.1
b*	18.7±0.0	60.5±1.6

Results are mean±SD of four individual determinations. FFA (free fatty acid). TPM (change in total polar material). CD (conjugated diene). AV (p-Anisidine value). L* (lightness/darkness). a* (redness/greenness). b* (yellowness/blueness color).

resulted in an increase in the measurement of all the aforementioned properties. Additionally, the viscosity of fresh oil at 30°C increased during frying and a* and b* values increased in the colour scale whereas the L* value decreased.

3.2 ADSORPTION USING A SINGLE ADSORBENT

The changes in used frying oil that were imposed by the use of different adsorbents during adsorption and the improvement ratios are provided in Table II. Statistically significant (*p*<0.05) differences were observed in terms of the different adsorbents used wherein the improvement ratios based on the parameters (FFA, TPM, CD content, AV, Hunter Lab colour values and viscosity) investigated.

Hydrolysis occurs during frying with the aid of heat and the effect of humidity from the food product itself and this process results in an increase in the FFA content of the oil. Therefore, it is of utmost importance to determine the FFA content [19]. The most effective adsorbent in reducing free acidity was AO as shown in Table II, wherein AO adsorbed FFA. Increasing the AO ratio from 3% to 6% reduced the acidity of the used frying oil from 0.85% to 0.60% then further down to 0.44%. The PI of FFA by AO increased from 37.3% to 61.2%. M reduced FFA down to 0.62%, when used at a ratio of 6% providing a PI of 34.3%. Other adsorbents such as SG, B, AC and Z also reduced FFA content to some extent. Moreover, increasing the SG or AC concentrations from 3% to 6% improved the removal of FFA.

Bhattacharya *et al.* [13] reported that the regeneration of two different samples of thermally polymerised palm oil using AO improved the FFA content by 36.50-38.24%, SG by 40.83-48.70%, AC by 39.38-39.97% and M by 52.39-58.59%. Kheang *et al.* [20] used SG, AO and acid activated BE in the regeneration of frying oil. The most effective among these adsorbents was SG that resulted in a decrease of 33.3% in FFA content and a 65.1% decrease in PV. Mono- and di-glycerides and polymer compounds are also formed in oil during frying. All these compounds constitute the polar fraction of oil and are considered as the non-triglyceride compounds [13]. SG was the most effective adsorbent in reducing the TPM content as indicated by the obtained results. SG facilitated the adsorption of mono- and diglycerides of polar character in addition to a partial adsorption of FFA. Increasing the SG content from 3% to 6% reduced the TPM from 10.5% first to 8.0% and further down to 6.5% at the higher concentration. The PI was 23.8% when 3% of SG was used and it was 38.1% when 6% of SG was used. BE and M succeeded also in the partial adsorption of polar materials, and increasing the adsorbent concentration decreased TPM content further with increasing PI. AO, which was effective in the removal of FFA, could only improve the TPM

content by 11.4% and 8.6% when it was used at concentrations of 3% and 6%, respectively.

Bhattacharya *et al.* [13] reported that the TPM content of used palm oil could be removed by 52.65-56.73% using SG, by 22.98-33.99% using M, by 21.32-22.84% using AC and by 28.33-34.20% by AO.

Hydroperoxides and CDs, which are the primary products of oxidation, resulted in an increase in absorbance at 232 nm in the UV range. In general, none of the adsorbents that were used could adsorb the primary products of oxidation at either concentration. Furthermore, thermal treatment at 60°C for 30 min further increased the CD content with respect to its value in the control sample.

In contrast to the case of the CDs, the AV, which is an indicator of the aldehyde content; the secondary products of oxidation, decreased when either adsorbent was used except for Z [19]. AC was the adsorbent that reduced the AV the most, improving by 62.7% when used at a concentration of 6%. SG, AO, B and BE at the same concentration provided improvements of more than 20%. M reduced the concentration of the secondary oxidation products by 29.0% when used at a concentration of 6%. Bhattacharya *et al.* [13] reported that the CD content and the AV of used palm oil could be decreased by 59.45-68.30% and 58.17-63.09% using M, by 36.10-43.52% and 33.94-35.13% using AC, by 38.83-47.12% and 23.95-32.31% using SG and by 67.85-68.10% and 17.80-20.23% using AO, respectively, each used at a concentration of 10%.

The viscosity of oils increased as a result of the formation of polymers during frying [19]. Some adsorbents succeeded in lowering the viscosity. However, increasing the concentration at which the adsorbent was used did not result in an improvement in the reducing of viscosity. SG was the adsorbent that provided the highest improvement (43%) when used at a concentration of 3%. The PI was 38.6% when 6% AC was used followed by 37.6% when 3% B was used. Bhattacharya *et al.* [13] reported an improvement of 13.09%-15.70% in the viscosity of used palm oil using 10% SG.

The addition of the adsorbent resulted in an increase in the L* Hunter colour value indicating a lightening of the colour and decrease in both a*, an indicator of the red-green colour, and b*, an indicator of the blue-yellow colour. BE, which is generally used in colour absorption of the raw oil during refinement, was able to provide the most extensive lightening of the colour attaining a difference of 17.2% when used at a concentration of 6%. The lowest a* (3.3) and b* (52.7) values were determined when 6% BE was used. This indicated that BE was effective in the removal of the red and the yellow colours. In addition to BE, AC was also effective in terms of colour change. Although AC lightened the colour and reduced the red and the yellow colour values, problems were encountered

Table II - Effect of single adsorbent treatment on chemical and physical properties of used frying oil

Adsorbent	FFA (%)		TPM (%)		CD (%)		AV (mmol/kg oil)		Viscosity (mPa.s, 30°C)		HunterLab color values			
	Value	PI	Value	PI	Value	PI	Value	PI	Value	PI	L*	a*	b*	TCD
UFO	0.85	-	10.5	-	0.54	-	34.4	-	64.2	-	46.9	14.6	60.5	-
AO (3%)	0.60±0.01h	37.3	9.3±0.0c	11.4	0.56±0.0cde	-8.0	27.0±6.0de	30.7	64.5±0.3c	-3.2	47.0±0.0m	14.4±0.0c	57.7±0.2g	2.9
BE (3%)	0.87±0.00a	-3.0	8.6±0.0e	18.1	0.57±0.01cd	-12.0	32.4±1.6abd	8.3	63.0±0.1d	12.1	54.0±0.0c	7.0±0.0m	55.0±0.1k	11.8
SG (3%)	0.74±0.02ef	16.4	8.0±0.0f	23.8	0.55±0.01cde	-4.0	28.5±0.9bcde	24.5	60.1±0.3h	43.0	49.7±0.0h	12.1±0.0g	57.9±0.0f	4.6
AC (3%)	0.83±0.01b	3.0	9.6±0.4bc	8.6	0.61±0.00ab	-28.0	27.2±0.3de	29.9	64.4±0.4c	-1.8	50.1±0.0g	9.1±0.0i	58.1±0.0f	6.8
Z (3%)	0.76±0.00cde	13.4	10.6±0.2a	-1.0	0.55±0.00de	-4.0	33.9±0.5ab	2.1	61.2±0.4f	31.4	47.5±0.0k	14.6±0.0b	57.2±0.1l	3.3
B (3%)	0.78±0.00cd	10.4	9.9±0.4b	5.7	0.55±0.02de	-4.0	27.2±4.2de	29.9	60.6±0.4g	37.6	48.3±0.0i	13.4±0.0e	59.4±0.1c	2.2
M (3%)	0.76±0.00cde	13.4	9.3±0.5cd	11.9	0.57±0.02cd	-12.0	33.4±0.8abc	4.1	65.6±0.1b	-14.8	51.9±0.0f	10.2±0.0h	60.5±0.1a	6.7
AO (6%)	0.44±0.02i	61.2	9.6±0.0bc	8.6	0.59±0.01bcd	-20.0	28.1±4.4bcde	26.1	64.1±0.0a	0.1	46.7±0.0n	14.3±0.0d	56.9±0.1i	3.7
BE (6%)	0.74±0.01fe	16.4	7.6±0.2f	28.1	0.52±0.03e	8.0	28.0±0.3bcde	26.6	62.5±0.3e	17.4	57.2±0.0a	3.3±0.0n	52.7±0.1l	17.2
SG (6%)	0.72±0.01fg	19.4	6.5±0.0g	38.1	0.56±0.02cde	-8.0	26.3±1.6e	33.6	63.2±0.4d	10.7	52.3±0.0e	9.3±0.0	58.5±0.0e	7.9
AC (6%)	0.71±0.00g	20.9	9.6±0.0bc	8.6	0.64±0.00a	-40.0	19.3±1.7f	62.7	60.5±0.3g	38.6	53.9±0.0d	7.1±0.0l	57.5±0.0h	10.7
Z (6%)	0.76±0.01de	13.4	10.7±0.2a	-1.9	0.59±0.04bc	-20.0	34.4±1.3a	0.0	65.3±0.0h	-11.5	47.4±0.0	14.7±0.0a	58.1±0.0f	2.5
B (6%)	0.79±0.00c	9.0	9.8±0.2bc	7.1	0.55±0.00de	-4.0	29.0±0.0abcde	22.4	61.1±0.4f	32.2	49.5±0.0	12.3±0.0f	59.9±0.0b	3.5
M (6%)	0.62±0.01h	34.3	8.8±0.2de	16.7	0.59±0.01bcd	-20.0	27.4±1.3cde	29.0	62.5±0.3e	18.0	54.0±0.0b	8.0±0.0k	59.2±0.0d	9.8

Results are mean±SD of two individual determinations. a-n Different letters in the same column for each group indicate a statistical difference between the mean values (*P*<0.05). UFO (used frying oil). AO (aluminum oxide). BE (bleaching earth). SG (silica gel). AC (activated charcoal). Z (zeolite). B (bentonite). M (magnesol XL). FFA (free fatty acid). TPM (change in total polar material). CD (conjugated diene). AV (*p*-Anisidine value). L* (lightness/darkness). a* (redness/greenness). b* (yellowness/blueness color). TCD (total color difference). PI (percentage improvement).

during filtration. The used oil needed to be centrifuged at 15000 rpm following two consecutive filtrations and colour determination could only be carried out afterward. Although AC was the most effective adsorbent in regaining the colour of the frying oil, since it also absorbs useful compounds such as tocopherols and that it is problematic in terms downstream filtration processing, it was unsuitable to be used as an adsorbent [2]. AC was very effective in the lightening of the colour of used palm oil, wherein an improvement of 83.63-94.73% in Lovibond red colour values and of 86.53-91.57% in yellow colour values was reported [13]. Among the commercial adsorbents, HB reduced the FFA content of oil by 84.5%, FP recovered the oil stability as much as 38.3% and M improved colour by 46.6% [14].

In summary, when used alone, AO was the most effective adsorbent in the removal of FFA, SG in reducing of the TPM content, AC in the removal of secondary oxidation products, BE in colour improvement and SG in reducing the viscosity.

3.3 ADSORPTION USING BINARY ADSORBENT MIXTURES

Using binary mixtures of the adsorbents was more effective than when the adsorbents were used alone. As shown in Table III, 6%AO+6%AC mixture was the most effective in reducing the FFA content, lowering the values from 0.85% down to 0.32% after 30 min of treatment. The PI was determined as 79.1%, wherein the PI of other mixtures varied between 11.9% and 67.2%. Increasing the AO content of the mixture from 3% to 6% further reduced FFA. For instance, the PI for the 3%AO+3%SG mixture was 50.7%, whereas it was improved to 67.2% in the 6%AO+3%SG mixture. The use of AO alone at a concentration of 6% resulted in a PI of 61.2%, whereas the addition of SG, BE, B or AC at a concentration of 3% resulted in PI values higher than 64%.

The binary mixtures with the lowest TPM content were 3%AO+6%SG and 3%M+3%Z with the value of 7.1%. The PI was calculated as 32.4%. Previous analyses indicated SG to be the single and the best adsorbent that lowered the TPM content. The TPM content of SG containing binary mixtures was below 8.4% with a PI above 21%. As it was the case for single adsorbents, M also facilitated the adsorption of polar materials. 3%SG+3%M reduced the TPM content down to 8.3% and 3%SG+6%M mixture reduced it to 8.0%. The lowest PI was observed at 5.7% in the 6%AO+6%Z mixture.

McNeill *et al.* [8] mixed different ratios of SG and AC with canola oil, stirred them in a water bath kept at 80°C for 20 min and filtered them following centrifugation. All binary adsorbent mixtures used were reported to lower the acidity value (51.1-53.9%), PV (39.4-53.5%), photometrical color values (38.3-49.7%), TPM content (16.3-27.5%), saturated (54.0-58.9%) and unsaturated (27.3-39.0%) carbonyl content.

Comparison of the CD contents of binary mixtures with the control samples displayed varying responses. The heat application during adsorption was thought to be effective in the formation of the CD. Some binary mixtures containing M were shown to be effective in the adsorption of the CD. The lowest CD content (0.31%) was recorded when 3%M+3%B mixture used to treat the oil, wherein the PI was 92.0%. This was followed by 3%MS+3%Z with a PI of 72%.

All adsorbent mixtures succeeded in reducing the AV when compared to the value of the control sample. The secondary products of oxidation were adsorbed by the adsorbent mixtures. The PI varied between 5.8% and 56.8%. The used frying oil had an AV of 34.4 mmol/kg of oil, whereas the oil treated with 6%AO+6%AC had an AV of 20.7 mmol/kg. AC was determined to be effective in the removal of secondary oxidation products when used alone. Therefore, AC containing mixtures were determined to lower the AV even further.

The binary mixture that reduced the viscosity the most (60.10 mPa.s) was 3%AO+6% SG. The presence of SG in the mixture was thought to be effective in lowering the viscosity. The performance of this mixture was followed by 3%SG+6%Z and 3%AL+3% MS mixtures. The use of single adsorbents or adsorbent mixtures did not yield considerable changes in the improvement of viscosity values.

The 3%ALO+6%BE mixture was the most successful in lightening the oil colour increasing the L* value from 46.9 to 57.5. The effect of BE performance was considerable in this mixture. This mixture also lowered the a* and b* values decreasing the intensity of red and yellow colours in oil. The total colour difference attained using this mixture was the highest (18.1). The lowest colour difference was detected when 6%AO+6%Z mixture was used. The effect of this mixture on lightening the colour of the oil was very low. Statistically, there were significant differences ($p<0.05$) in the parameters measured prior to and following treatment with binary adsorbent mixtures. Bhattacharya *et al.* [13] reported 2.5% aluminium hydroxide + 7.5% AC mixture and the 7.5% M + 2.5% SG mixture to be the most effective ones for treating palm oil in batch systems among the binary mixtures investigated. They reported SG and M to perform better in binary mixtures than being used alone in improving the physical and chemical characteristics of the oil.

3.4 ADSORPTION USING TERNARY ADSORBENT MIXTURES

The most effective ternary mixture in lowering FFA was 3%AO+3%BE+3%Z providing an improvement

Table III - Effect of double adsorbent treatment on chemical and physical properties of used frying oil

Adsorbent mixture	FFA (%)		TPM (%)		CD (%)		AV (mmol/kg oil)		Viscosity (mPa.s, 30°C)		HunterLab color values			TCD
	Value	PI	Value	PI	Value	PI	Value	PI	Value	PI	L*	a*	b*	
UFO	0.85	-	10.5	-	0.54	-	34.4	-	64.2	-	46.9	14.6	60.5	-
3%AO+3%BE	0.54±0.00cdef	46.3	8.0 ±0.0efg	23.8	0.53±0.02defg	4.0	30.1±0.6abc	17.8	62.8±0.1de	15.1	54.3±0.0e	6.6±0.0r	55.1±0.0i	12.2
3%AO+6%BE	0.54±0.04ef	46.3	7.6±0.2g	28.1	0.49±0.01g	20.0	31.2±1.4ab	13.3	61.6±0.3g	27.1	57.5±0.0a	2.9±0.0v	51.7±0.0m	18.1
3%AO+3%SG	0.51±0.00fg	50.7	7.6±0.2g	28.1	0.51±0.00efg	12.0	30.2±1.5abcd	17.4	63.6±0.3b	6.3	49.6±0.0n	11.7±0.0f	57.4±0.3g	5.1
3%AO+6%SG	0.51±0.07fg	50.7	7.1±0.0h	32.4	0.50±0.01fg	16.0	25.8±1.4de	35.7	60.1±0.3i	42.6	52.3±0.0h	9.1±0.0k	57.4±0.0g	8.3
6%AO+3%BE	0.41±0.02h	65.7	7.6±0.2g	28.1	0.51±0.01efg	12.0	28.4±0.0bcd	24.9	62.3±0.2ef	19.8	54.6±0.0c	6.2±0.0s	54.7±0.0k	12.8
6%AO+3%SG	0.40±0.02h	67.2	8.3±0.0de	21.0	0.53±0.03defg	4.0	27.4±0.6cde	29.0	62.4±0.2def	18.2	49.0±0.0o	11.9±0.0e	57.4±0.0g	4.6
3%AO+3%AC	0.54±0.01def	46.3	9.5±0.2bc	10.0	0.55±0.01cde	-4.0	24.7±0.7e	40.2	63.9±0.0a	-4.0	49.8±0.0m	8.6±0.0ll	56.6±0.1i	7.8
6%AO+6%Z	0.49±0.02g	53.7	9.9±0.4af	5.7	0.54±0.02cdef	0.0	30.4±0.1abc	16.6	63.4±1.2bc	8.2	47.7±0.0t	14.3±0.0a	59.2±0.1c	1.5
3%SG+3%Z	0.77±0.04a	11.9	7.7±0.0fg	26.7	0.56±0.01cd	-8.0	28.9±1.7bc	22.8	64.7±0.4a	-5.3	48.7±0.0p	13.4±0.0c	56.4±0.0l	4.7
3%SG+6%Z	0.71±0.00b	20.9	8.0±0.0e	23.8	0.53±0.01cdefg	4.0	27.9±0.3bcd	27.0	60.9±0.4h	34.5	48.5±0.0r	13.5±0.0b	58.6±0.0d	2.7
6%AO+3%B	0.42±0.05h	64.2	9.5±0.2bc	10.0	0.59±0.00bc	-20.0	33.0±2.5a	5.8	63.4±0.2bc	8.5	48.0±0.0s	13.1±0.0d	57.8±0.1f	3.3
3%SG+3%B	0.76±0.00ab	13.4	8.0±0.0efg	23.8	0.56±0.01cde	-8.0	28.1±0.2bcd	26.1	63.6±0.1b	5.7	52.1±0.1l	9.6±0.0	59.1±0.2c	7.4
3%SG+6%B	0.76±0.00ab	13.4	7.7±0.0fg	26.7	0.68±0.08a	-56.0	26.5±0.3de	32.8	62.8±0.3de	14.7	53.1±0.0g	8.5±0.0m	58.3±0.1e	9.0
3%SG+3%M	0.64±0.02c	31.3	8.2±0.2ef	22.4	0.58±0.01cd	-16.0	28.4±1.9bcd	24.9	62.7±0.1de	15.9	54.4±0.0d	7.3±0.0p	59.0±0.2c	10.6
3%AO+3%M	0.45±0.00gh	59.7	9.6±0.0ab	8.6	0.56±0.01cde	-8.0	30.6±0.2ab	15.8	60.8±0.4h	35.0	52.4±0.0h	9.6±0.0	59.0±0.1c	7.5
3%SG+6%M	0.58±0.02cde	40.3	8.0±0.0efg	23.8	0.54±0.00cdef	0.0	26.6±0.1de	32.4	62.0±0.3fg	23.0	55.6±0.0b	6.0±0.0t	57.1±0.0h	12.7
3%AO+6%M	0.43±0.00h	62.7	8.6±0.0d	18.1	0.38±0.02h	64.0	28.9±0.5bcd	22.8	62.5±0.1def	17.7	53.8±0.0f	8.1±0.0h	60.8±0.0a	9.5
6%AO+6%AC	0.32±0.02i	79.1	8.8±0.2c	16.7	0.63±0.00b	-36.0	20.7±0.0f	56.8	62.5±0.3def	17.4	51.9±0.0i	5.9±0.0u	54.4±0.2k	11.7
6%AO+3%AC	0.41±0.02h	65.7	9.1±0.3c	13.3	0.39±0.02h	60.0	24.3±0.1e	41.9	61.7±0.3g	26.1	47.1±0.0u	7.7±0.0o	53.5±0.1l	9.9
3%M+3%Z	0.60±0.04cd	37.3	7.1±0.4h	32.4	0.36±0.04h	72.0	26.3±0.9de	33.6	62.9±0.2cd	13.0	51.1±0.0l	11.0±0.0g	60.3±0.1b	5.5
3%M+3%B	0.75±0.02ab	14.9	8.3±0.0de	21.0	0.31±0.01i	92.0	29.9±4.1abc	18.7	63.5±0.1b	7.2	51.4±0.0k	10.7±0.0h	58.7±0.1d	6.2

Results are mean±SD of two individual determinations. a-u Different letters in the same column for each group indicate a statistical difference between the mean values (P<0.05). UFO (used frying oil). AO (aluminum oxide). BE (bleaching earth). SG (silica gel). AC (activated charcoal). Z (zeolite). B (bentonite). M (magnesol XL). FFA (free fatty acid). TPM (change in total polar material). CD (conjugated diene). AV (p-Anisidine value). L* (lightness/darkness). a* (redness/greenness). b* (yellowness/blueness color). TCD (total color difference). PI (percentage improvement).

Table IV - Effect of triple adsorbent treatment on chemical and physical properties of used frying oil

Adsorbent mixture	FFA (%)		TPM (%)		CD (%)		AV (mmol/kg oil)		Viscosity (mPa.s, 30°C)		HunterLab color values			
	Value	PI	Value	PI	Value	PI	Value	PI	Value	PI	L*	a*	b*	TCD
UFO	0.85	-	10.5	-	0.54	-	34.4	-	64.2	-	46.9	14.6	60.5	-
3%AO+3%SG+3%Z	0.51±0.00bc	50.7	7.0±0.2ef	33.8	0.61±0.03ab	-28.0	28.3±0.4bc	25.3	62.3±0.4ef	19.9	48.4±0.0r	13.3±0.0a	59.5±0.0c	2.3
3%AO+3%BE+3%Z	0.33±0.00h	77.6	7.1±0.0e	32.4	0.56±0.02ab	-8.0	25.7±0.1bcde	36.1	60.7±0.2bhi	35.9	51.2±0.0n	10.0±0.0d	55.2±0.0l	8.3
6%AO+3%BE+3%Z	0.51±0.00bc	50.7	6.8±0.0ef	35.2	0.52±0.00ab	8.0	26.3±0.7bcd	33.6	63.5±0.2cb	7.3	51.7±0.0h	9.1±0.0h	55.8±0.1k	8.7
3%AO+6%BE+3%Z	0.51±0.00bc	50.7	6.5±0.0fg	38.1	0.53±0.02ab	4.0	20.1±1.9f	59.3	61.6±0.4g	26.7	55.1±0.1b	5.3±0.0o	51.7±0.2n	15.2
3%AO+6%SG+3%Z	0.47±0.02de	56.7	6.2±0.0g	41.0	0.51±0.04b	12.0	25.1±0.9bcdef	38.6	63.0±0.3de	12.9	50.2±0.0o	11.4±0.0c	58.4±0.0e	5.0
3%AO+3%BE+3%AC	0.48±0.00cde	55.2	9.3±0.0b	11.4	0.56±0.02ab	-8.0	26.0±0.8bcd	34.9	61.1±0.4h	32.3	56.7±0.1a	3.7±0.0p	53.3±0.1m	16.4
3%AO+3%SG+3%AC	0.5±0.00cd	52.2	7.7±0.0d	26.7	0.56±0.00ab	-8.0	23.9±0.7cdef	43.6	59.5±0.3j	48.4	54.0±0.0c	5.7±0.0n	55.6±0.2k	12.4
3%AO+6%AC+3%Z	0.46±0.02def	58.2	8.6±0.0c	18.1	0.56±0.03ab	-8.0	20.6±0.0ef	57.3	63.2±0.3cd	10.7	53.3±0.0f	6.6±0.0m	56.9±0.2j	10.9
6%AO+3%AC+3%Z	0.42±0.00g	64.2	8.2±0.2c	22.4	0.53±0.01ab	4.0	23.8±1.4cdef	44.0	62.1±0.4fg	21.6	52.4±0.0h	8.6±0.0l	58.7±0.0d	8.3
3%AO+3%SG+3%B	0.49±0.02cd	53.7	7.6±0.2d	28.1	0.62±0.16a	-32.0	29.9±1.0ab	18.7	59.6±0.3j	47.6	51.8±0.0	9.8±0.0e	57.3±0.1h	7.5
6%AO+3%SG+3%B	0.42±0.02fg	64.2	7.6±0.2d	28.1	0.52±0.03b	8.0	28.4±0.3bc	24.9	61.7±0.4g	25.8	51.3±0.0m	9.8±0.0e	57.6±0.0g	7.1
3%AO+3%AC+3%B	0.44±0.02efg	61.2	8.6±0.0c	18.1	0.59±0.01ab	-20.0	23.3±0.9cdef	46.1	60.3±0.3i	40.0	52.2±0.0	8.1±0.0j	57.1±0.0l	9.1
3%AO+3%AC+6%B	0.46±0.03def	58.2	8.5±0.2c	19.5	0.57±0.00ab	-12.0	24.1±0.0cdef	42.7	63.1±0.4cd	11.5	53.4±0.0e	7.2±0.0l	57.9±0.1f	10.2
3%AO+3%M+3%Z	0.42±0.00fg	64.2	7.0±0.2ef	33.8	0.39±0.03c	60.0	19.3±1.9b	62.7	63.0±0.1a	12.5	52.3±0.0	9.7±0.0f	58.6±0.0ed	7.6
6%AO+3%SG+3%Z	0.51±0.04bc	50.7	6.7±0.2ef	36.2	0.36±0.01dc	72.0	21.1±7.7def	55.2	63.3±0.4c	9.1	48.7±0.0p	13.0±0.0b	58.6±0.0d	3.1
3%AO+3%M+3%B	0.49±0.02cd	53.7	9.3±0.5b	11.9	0.37±0.03dc	68.0	33.9±1.4a	2.1	63.8±0.2b	9.2	52.9±0.0g	9.1±0.0h	59.9±0.3b	8.2
3%AO+3%AC+6%Z	0.55±0.02b	44.8	8.6±0.0c	18.1	0.30±0.01dc	96.0	23.7±3.1cdef	44.4	61.0±0.3h	33.7	52.0±0.0k	9.4±0.0g	60.6±0.1a	7.3
3%SG+3%M+3%B	0.64±0.02a	31.3	9.9±0.4a	5.7	0.29±0.00d	100.0	30.2±0.9ab	17.4	61.8±0.2g	25.3	54.0±0.0d	7.5±0.0k	59.6±0.1c	10.1

Results are mean±SD of two individual determinations. a-r Different letters in the same column for each group indicate a statistical difference between the mean values (P<0.05). UFO (used frying oil). AO (aluminum oxide). BE (bleaching earth). SG (silica gel). AC (activated charcoal). Z (zeolite). B (bentonite). M (magnesol XL). FFA (free fatty acid). TPM (change in total polar material). CD (conjugated diene). AV (p-Anisidine value). L* (lightness/darkness). a* (redness/greenness). b* (yellowness/blueness color). TCD (total color difference). PI (percentage improvement).

of 77.6% (Tab. IV). This mixture was followed by 3%AO+3%SG+3%B, 6%AO+3%AC+3%Z and 3%ALO+3%M+3%Z. The PI of these mixtures was calculated as 64.2%. All ternary mixtures were more successful in improving the FFA content of oil than the binary mixtures. The ternary mixture that best reduced the TPM was 3%AO+6%BE+3%Z which reduced the value of 10.5% in control samples down to 6.2%. The PI for this ternary mixture was determined as 41.0%. The presence of SG at a concentration of 6% in the mixture further reduced the TPM content. The fluctuation was observed in CD contents in comparison to the control samples. Specifically, M containing ternary mixtures succeeded in the adsorption of the CD. The CD improvement ratio for 3%SG+3%M+3%B, 3%AO+3%AC+6%Z, 3%ALO+3%M+3%B and 3%AO+3%M+3%Z mixtures were determined as 100%, 96.0%, 68.0% and 60.0%, respectively.

All AV were lower than their control counterparts. Both binary and ternary adsorbent mixtures succeeded in the removal of secondary products of oxidation (aldehydes). The highest decrease was attained in the 3%AO+3%M+3%Z mixture. The AV decreased from 34.4 to 19.3. The PI was 62.7%. The AC containing mixtures were also successful in reducing the AV. The mixtures that best lowering the viscosity were 3%AO+3%SG+3%AC (48.4%) and 3%AO+3%SG+3%B (47.6%). The SG in the mixtures facilitated the reducing of the viscosity. Generally, both the binary and the ternary mixtures succeeded in lowering the viscosity.

The highest colour difference in ternary mixtures was attained as 16.3 when the 3%AO+3%BE+3%AC mixture was used. The BE and the AC facilitated the lightening of the colour. The mixture containing 6%BE; 3%ALO+6%BE+3%Z, achieved 15.2% difference in total colour. The ternary adsorbent mixtures resulted in the increase in L* value and the reducing of the a* and b* values. As in the case of using a single type of adsorbents or their utilisation as binary mixtures, the ternary mixtures affected the indicated parameters of used oil samples in a statistically significant manner ($p<0.05$).

Lin et al. [21] mentioned that the 3%HB+3%M+2% FP and the 2%HB+3%M+2% BR mixtures were most effective in the regeneration of used oil. The optimum mixing time was determined as 6-9 min, whereas a reduction of 82.6% and 87.6% in FFA content, 26.8% and 32.6% in colour absorbance values as well as 5.6% and 8.6% in oil sensor readings was observed for the mixtures, respectively.

Several studies reported that the use of adsorbents during deep-frying could increase the active usage period of the oil. Mancini-Filho et al. [12] reported that the addition of several adsorbents (bleaching clay, AC, magnesium oxide, celite and their mixtures) or additives (ascorbyl palmitate) to the shortenings used during deep-frying increased the stability of

the oil. Maskan and Bağcı [22] and Maskan [18] reported that during repeated deep fat frying, adding the 2%pekmez earth+3%B+3%magnesium silicate mixture into the oil of pre-determined volume reduced the free acidity, CD content, PV, viscosity and Hunter a* and b* values of the sunflower seed oil after 50 rounds of deep-frying.

4. CONCLUSION

The study showed that adding adsorbents singly or in binary and ternary mixtures to used frying oil during adsorption generally reduced the FFA content, TPM content and AV, whereas it increased L* and reduced a* and b* colour values. Some adsorbents and their mixtures resulted in either a decrease or an increase in viscosity and CD contents. Binary and ternary mixtures were more effective in reducing the FFA in comparison to the use of a single adsorbent. Additionally, some adsorbents, although performed poorly in reducing the polar material content, performed successfully in binary and ternary mixtures. Binary and ternary adsorbent mixtures performed better in the adsorption of the primary and secondary products of oxidation than the single adsorbent did. Adsorbents used in this study would be suitable for use either singly or in binary and ternary mixtures in the regeneration of used oil.

Compliance with ethical standards
Conflict of interest: there are no conflicts of interest to declare.
Compliance with ethics requirements: this article does not contain any studies with human or animal subjects.

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